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## **Supporting Information**

## Scalable, low-cost synthesis of high volumetric capacity LiMn<sub>0.5</sub>Fe<sub>0.5</sub>PO<sub>4</sub> cathode for lithium-ion batteries

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**Fig. S-1**. Two-step carbon coating synthesis process for S-LMFP. LMFO is synthesized and mixed with the additional precursors for LMFP and calcined to form an intermediate product. The intermediate product is mixed with more carbon and fed into the spray dryer as an aqueous solution, which upon calcination forms dense LMFP.



**Fig. S-2.** Mixed solution of  $Fe_{0.5}Mn_{0.5}O$ ,  $NH_4H_2PO_4$ , and  $H_2O$ . The high viscosity indicates  $Mn^{2+}$  dissolution has caused byproducts to form that inherently affect the solution rheology.



Fig. S-3. (a) SEM image of  $NH_4Mn_{0.5}Fe_{0.5}PO_4$  with EDS mapping of (b) Fe and (c) Mn.



**Fig. S-4**. XRD patterns of pristine LMFO and LMFO doped with 1 mol% Mg and 1 mol% Co.

Sample	a (Å)	b (Å)	c (Å)	Crystal Size (nm)	$R_{wp}$
S-LMFP	4.72	10.40	6.05	152	10.2%
P-LMFP	4.72	10.38	6.05	151	10.0%

**Table S-1**. Lattice parameters and crystallite size values obtained from Rietveld refinement of

the XRD patterns of S-LMFP and P-LMFP cathodes



Fig. S-5. Low magnification SEM images of (a) S-LMFP, (b) P-LMFP, and (c) O-LMFP.



**Fig. S-6**. Electrochemical performances of all three LMFP samples on a gravimetric basis. (a) Half-cell cycling performances of all three LMFP samples and (b) voltage profiles recorded at C/5 charge and discharge rates. Aerial capacities of all the samples with 1.4 to 1.6 mA h cm<sup>-2</sup>. The press densities of S-LMFP, P-LMFP, and O-LMFP are, respectively, 2.31, 1.93, and 2.40 g cm<sup>-3</sup>.